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Part I

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THERMAL CONDUCTIVITY OF ANISOTROPIC SOLIDS AT HIGH TEMPERATURES

The Thermal Conductivity of Molded and Pyrolytic Graphites

Part I

TECHNICAL DOCUMENTARY REPORT NO. ASD-TDR-62-608

November 1962

Directorate of Materials and Processes
Aeronautical Systems Division
Air Force Systems Command
Wright-Patterson Air Force Base, Ohio

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Project No. 7364, Task No. 73652

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(Prepared under Contract No. AF 33(616)-7123
by the University of Cincinnati, Cincinnati, Ohio;
Michael Hoch, Joseph Vardi, authors)

1. Heat transfer
2. Graphite
- I. AFSC Project 7364,
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- II. Contract AF 33
(616)-7123
- III. University of
Cincinnati, Ohio
- IV. M. Hoch, J. Vardi
- V. Avail fr OTS
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DUCTIVITY OF ANISOTROPIC SOLIDS AT HIGH TEM-
PERATURES: The Thermal Conductivity of Molded
and Pyrolytic Graphites. Interim report, Nov
62, 19 pp incl illus., tables, 12 refs.

Unclassified Report

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dimensional, steady-state heat conduction in
a cylinder of finite length heated in vacuum

(over)

by high frequency induction and radiating
heat to the surroundings. The method has
been used to determine the radial thermal
conductivity, k_r , and the axial thermal con-
ductivity, k_z , of molded ZT type and pyro-
lytic graphite in the temperature range
1200°-2200°K. For ZT type graphite $k_z/k_r =$
 $-0.10116 + 2.00191 \times 10^{-4} \times T$ (1260°K < T
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0.0376 at 1817°K.

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FOREWORD

This report was prepared by Michael Hoch and Joseph Vardi, of the University of Cincinnati, under Contract No. AF 33(616)-7123. This research was carried out under Project No. 7364, "Experimental Techniques for Materials Research," Task No. 73652, "Intense Thermal Energy Transferred to Materials." The work was administered under the direction of the Directorate of Materials and Processes, Deputy Commander/Technology, Aeronautical Systems Division, with Mr. Hyman Marcus acting as Project Engineer.

This report covers work conducted from February, 1961, to February, 1962.

ABSTRACT

A method has been developed for the determination of the thermal conductivities of anisotropic solids under conditions of two-dimensional, steady-state heat conduction in a cylinder of finite length heated in vacuum by high frequency induction and radiating heat to the surroundings. The method has been used to determine the radial thermal conductivity, k_r , and the axial thermal conductivity, k_z , of molded ZT type and pyrolytic graphite in the temperature range 1200-2200°K. For ZT type graphite $k_z/k_r = -0.10116 + 2.00191 \times 10^{-4} \times T$ (1260°K < T < 2199°K); for pyrolytic graphite, $k_z/k_r = 0.0376$ at 1817°K.

This technical documentary report has been reviewed and is approved.



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NOMENCLATURE

a	radius of the specimen in cm
A	a constant
J_0	a zero order Bessel function of the first kind
J_1	a first order Bessel function of the first kind
J_2	a second order Bessel function of the first kind
k	thermal conductivity in cal/cm-sec-°K
k_n	thermal conductivity in the n direction in cal/cm-sec-°K
k_r	thermal conductivity in the r direction in cal/cm-sec-°K
k_z	thermal conductivity in the z direction in cal/cm-sec-°K
L	half height of the specimen in cm
n	an integer positive number 1, 2, 3
r	independent variable for radius in cylindrical coordinates
T	dependent variable for temperature in °K
T_a	temperature $T(a,1)$ in °K
T_0	temperature $T(0,1)$ in °K
T_s	constant surface temperature in °K
ΔT	temperature difference $T(a,1) - T(0,1)$ in °K
ΔT_r	temperature difference $T(r,0) - T(0,1)$ in °K
T'	dependent variable for temperature in °K, the contribution of the complementary solution

w	$\sqrt{k_z/k_r}$
x	an independent variable in a Cartesian Coordinate System
y	an independent variable in a Cartesian Coordinate System
z	independent variable for distance in cylindrical and Cartesian coordinates
α	the slope of the equation $T = T_0 + \alpha r^2$ in $^{\circ}\text{K}/\text{cm}^2$
β	constant, equal λ_n/a
ϵ	the total emissivity
ϵ_λ	spectral emissivity at a wavelength λ
λ_n	the respective zeros of a zero order Bessel function $J_0(\lambda_n) = 0$
θ	independent variable for angle in cylindrical coordinates
τ	dependent temperature equal to $T + T'$ in $^{\circ}\text{K}$
σ	Stephan Boltzmann radiation constant
ψ_0	function $\sum_{n=1}^{\infty} \tanh\left(\frac{\lambda_n L}{wa}\right) \frac{J_2(\lambda_n)}{\lambda_n J_1^2(\lambda_n)}$

INTRODUCTION

This study deals with solids having identical properties along the x and y directions and different properties along the z direction, where x, y, and z are the axes of a Cartesian Coordinate system. The aim is to determine the thermal conductivity in the x and y directions, which will be designated by k_x , and the thermal conductivity in the z direction, which will be designated by k_z .

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ANALYSIS

Using the physical model described earlier¹, the following partial differential equation may be derived for a cylindrical specimen whose axis coincides with the z direction of the anisotropy of the solid.

$$\frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r} + w^2 \frac{\partial^2 T}{\partial z^2} = 0 \quad (1)$$

where $w^2 = k_z/k_r$. Equation (1) assumes constant value of k_z and k_r in the temperature range of the measurement. The earlier case¹ is a special case of equation (1) if w is set equal to 1. With the appropriate initial conditions applied to the physical model, it may be shown that equation (1) may be satisfied by

$$T(r,z) = T_s - AJ_0(\beta r) \cosh(\beta z/w) \quad (2)$$

Two boundary conditions are required to obtain the temperature distribution of the specimen for the evaluation of the thermal conductivities.

A natural boundary condition that can be used is the expression in equation (3), obtained by equating the heat conducted into a small element of area on the flat end surface of the specimen to the heat radiated from the same element of area.

$$k_z = - \frac{\sigma \epsilon T^4}{\partial T(r,L)/\partial z} = - \frac{\sigma \epsilon T^4}{(\partial T/\partial z)_{r,L}} \quad (3)$$

Similar to the earlier case¹, equation (3) produces a non-linear boundary value problem which is unsolvable with common procedures. Therefore the empirical expression in equation (4) for the temperature at the flat end surface and the approximation in equation (5) of the temperature of the cylindrical surface as isothermal were applied to equation (2) to produce the temperature distribution within the solid expressed by equation (6).

$$T(r,L) = T_0 + \alpha r^2 \quad (4)$$

$$T(a,z) = T_s \quad (5)$$

$$T(r,z) = T_s - 4\alpha a^2 \sum_{n=1}^{\infty} \frac{\cosh(\lambda_n z/wa) J_0(\lambda_n)}{\cosh(\lambda_n L/wa) \lambda_n^2 J_1^2(\lambda_n)} J_0(\lambda_n r/a) \quad (6)$$

By differentiating equation (6), term by term, the temperature gradient in the z direction is obtained. The gradient at $r = 0$ and $z = L$ is

$$\left(\frac{\partial T}{\partial z}\right)_{0,L} = - \frac{4\alpha a}{w} \sum_{n=1}^{\infty} \tanh(\lambda_n L/wa) \frac{J_2(\lambda_n)}{\lambda_n J_1^2(\lambda_n)} = - \frac{4\alpha a}{w} \psi_0(L/wa) \quad (7)$$

Combining equations (7) and (3)

$$k_z = \frac{\sigma \epsilon T_0^4}{(\partial T / \partial z)_{0,L}} = \frac{\sigma \epsilon T_0^4}{\frac{4\alpha a}{w} \psi_0} \quad (8)$$

Using $w = \sqrt{k_z/k_r}$ and $\alpha a^2 = \Delta T$, equation (8) yields

$$\alpha a \psi_0 = \frac{\Delta T}{a} \psi_0 = \sigma \epsilon T_0^4 / 4 \sqrt{k_r k_z} \quad (9)$$

All quantities in equation (9) are known or measurable experimentally except the thermal conductivities k_r and k_z . Two equations of the form of equation (9) are sufficient to solve for k_z and k_r ; such equations are obtained by taking experimental data at a temperature T_0 for two different specimens of the same material with different L/a (preferentially same a). The right hand side of equation (9) is the same for both specimens; thus if the left hand side is plotted against w for each sample, the point of intersection of the two curves determines the correct values of k_r and k_z . The abscissa determines $w (= \sqrt{k_z/k_r})$ and the ordinate the value of the quantity $\sigma \epsilon T_0^4 / 4 \sqrt{k_r k_z}$. Values of the function ψ_0 are identical with the value of the constant K_0 calculated earlier¹ if the abscissa is changed from L/a to the variable L/aw .

Reexamination of equation (6) shows that $(\partial T / \partial z)_{a,z}$ is equal to zero, which yields, via equation (3), a value

approaching infinity for k_z . The value of k_z should be constant and independent of the point of evaluation. Thus an error is introduced by the boundary condition in equation (5) which defines the temperature on the cylindrical surface as isothermal and forces $(\partial T/\partial z)_{a,z}$ to be zero. An estimate of the error inherent in the solution of equation (9) can be obtained by replacing equation (6) by a perturbed boundary condition solution. The new boundary condition is chosen such that it forces k_z , evaluated at $r = a$ and $z = L$, to equal k_z , evaluated at $r = 0$ and $z = L$. This is accomplished by selecting an appropriate value for $(\partial T'/\partial z)_{a,L}$, where T' denotes the contribution of the complementary solution added to equation (7). The new solution may be written

$$\mathcal{T}(r,z) = T(r,z) + T'(r,z) \quad (10)$$

Replacing $T(r,z)$ by $\mathcal{T}(r,z)$ in equation (3) yields

$$k_z = - \frac{\sigma \epsilon T_0^4}{(\partial \mathcal{T}/\partial z)_{0,L}} = \frac{\sigma \epsilon T_0^4}{(\partial T/\partial z)_{0,L} + (\partial T'/\partial z)_{0,L}} \quad (11)$$

A comparison of equation (11) with equation (8) shows that the ratio of $(\partial T'/\partial z)_{0,L}$ to $(\partial T/\partial z)_{0,L}$ will estimate the error inherent in the solution of equation (9). Calculation of the term $(\partial T'/\partial z)_{0,L}$ with the assumption that the isothermal temperature on the cylindrical surface is correct within

10% of ΔT has been carried out earlier¹ for the case of an isotropic solid. Similar analysis for the anisotropic case shows that with a selection of $L/wa \leq 1$ (by choosing appropriate value of L/a), the term $(\partial T'/\partial z)_{0,L}$ can be neglected and the solution presented in equation (9) evaluates the thermal conductivity within an error of less than 6%. For $L/wa \leq 0.1$ the error is practically zero.

EQUIPMENT, EXPERIMENTAL PROCEDURE, AND MATERIALS

The equipment used was similar to that described earlier²

The previously described experimental procedure¹ was modified for the measurements. Cylindrical specimens of different ratios of length to diameter were heated with high frequency induction. The temperature gradients at different radial distances on the flat end surface of the cylinder were measured.

The molded graphite, ZT type, was obtained from the National Carbon Co. Its properties, as determined by the company are as follows:

Sample	Density gm/cm ³	Anisotropy
		Ratio of electrical resistivity (z direction over r direction) at room temperature
G-3A	1.980	2.50
G-7	1.978	2.50
G-5	2.000	2.86
G-9	2.000	2.86

The pyrolytic graphite was obtained from the General Electric Co. and was made at their Detroit plant (Sample P-3 was produced in their run No. 294).

EXPERIMENTAL RESULTS

The detailed experimental measurements on one sample (G-5) at one temperature are given in Table I. Similar measurements were taken at other temperatures and on other samples. ΔT is obtained by drawing a line through the average values of ΔT_r and $(r/a)^2$ and the origin ($\Delta T_r = 0$, $(r/a)^2 = 0$). The data in Table I show the correctness of equation (4).

Table II shows the experimental measurements on sample P-3, the best pyrolytic graphite. The sample is obviously not quite circularly symmetric, as ΔT_r values on one diameter, but on opposite sides (denoted + and ++) of the center, are different. Some samples tested showed negative ΔT_r 's and obviously had to be discarded.

Table III gives summarized results of measurements on two sets of ZT type graphite specimens (G3A-G7, and G5-G9). Table IV gives data on pyrolytic graphite.

Figure 1 shows the procedure of obtaining the thermal conductivities from the ΔT values for samples G-5 and G-9 at $T_0 = 1647^\circ\text{K}$. The lines are obtained as follows: values of the function $\psi_0(L/aw)$ for different values of L/aw are selected; the corresponding w values for each sample and the expression $\frac{(\Delta T/a)_{G-9}}{(\Delta T/a)_{G-5}} \psi_0$ are calculated for each value of

L/aw. The G-5 line is a plot of ψ_0 vs. w_{G-5} and the G-9 line is a plot of $\frac{(\Delta T/a)_{G-9}}{(\Delta T/a)_{G-5}} \psi_0$ vs. w_{G-9} . The point of intersection of the two lines is $w = 0.462$;

$$\frac{1}{(\Delta T/a)_{G-5}} \frac{\sigma \epsilon T_0^4}{\sqrt[4]{k_r k_z}} = 0.438 \text{ which represents two equations with}$$

the unknowns k_z and k_r . The simultaneous solution of the two equations gives the k values shown in Table III.

The thermal conductivities are presented in Tables III and IV in terms of the total emissivity of the flat surface of the specimen. The ratio k_z/k_r is independent of the value of the total emissivity. In the last two columns in each table k_r and k_z are evaluated by assuming gray bodies, $\epsilon = \epsilon_\lambda$.

DISCUSSION

An error in the thermal conductivity values is introduced because of the mathematical approximation. This error is a function of the term L/aw . A selection of a small value for L/aw reduces the error; however, other factors such as the experimentally desirable value for ΔT must also be considered. The mathematical error introduced for samples G-5 ($L/aw = 0.53$) and G-9 ($L/aw = .247$), for G-3A ($L/aw = 0.888$) and G-7 ($L/aw = 0.226$), for P-3 ($L/aw = 0.89$) and P-3A ($L/aw = 0.236$), are 5%, 6%, and 6% respectively.

Errors may also be introduced by the experimental measurements of the temperature. The values of the ΔT_r terms given in Table I are evaluated from measurements of $T(r,L)$. The radial distances r are along two perpendicular diameters. On one diameter r is $1a, 3/4a, 1/2a, 1/4a, 0, 1/4a, 1/2a, 3/4a, 1a$; on the other, $1a, 2/3a, 1/3a, 0, 1/3a, 2/3a, 1a$. Such points are useful for checking the empirical boundary condition in equation (4). This is essential in the case of pyrolytic graphite: pyrolytic graphite should exhibit the kind of anisotropy considered here³. The layers in the x and y directions are not completely parallel to each other and not perfectly perpendicular to the z direction^{4,5}. If the layers are not parallel and perpendicular to the z axis, the angular symmetry $\partial T / \partial \theta = 0$ does not hold, and the method cannot be

applied. Thirty samples of pyrolytic graphite were tested, and none was completely symmetrical. Samples P-1 and P-2 showed less deviation than the others. Sample P-3 was the most symmetrical; it was cut down in height and reused as sample P-3A.

To correct the measured temperatures for non-black body conditions it was necessary to obtain values of the spectral emissivities. The spectral emissivities (for polished surface) were measured and found to be $\epsilon_{0.665} = 0.61064 - 1.85699 \times 10^{-4} \times T$ (for $1265 < T < 2195^\circ\text{K}$) for the ZT type graphite and $\epsilon_{0.665} = 0.262$ at 1875°K for the pyrolytic graphite. It is estimated that the accuracy in the measurement of the temperature is $\pm 3^\circ\text{K}$. For the ZT type graphite (samples G-5 and G-9 at 1647°K) the standard deviations are 1.8%, 2.1%, and 2.8% for $(\Delta T)_{G-5}$, $(\Delta T)_{G-9}$, and $(\Delta T)_{G-5}/(\Delta T)_{G-9}$, respectively. For the pyrolytic graphite (samples P-1 and P-2 at 1808°K) the standard deviations are 10.6%, 6.6%, and 12.5% for $(\Delta T)_{P-1}$, $(\Delta T)_{P-2}$, and $(\Delta T)_{P-1}/(\Delta T)_{P-2}$, respectively. The above error in $(\Delta T)_{P-1}/(\Delta T)_{P-2}$ produces a standard deviation of 68.9% in the ratio k_z/k_r ; for the samples P-3 and P-3A at 1817°K the standard deviation is 2.69%, 2.12%, and 3.0% for $(\Delta T)_{P-3}$, $(\Delta T)_{P-3A}$, and $(\Delta T)_{P-3}/(\Delta T)_{P-3A}$, respectively, which produces a standard deviation of 5.8% for the ratio k_z/k_r . Taking a certainty

level of 95%, the experimental error in k_z/k_r for the samples P-1 and P-2 is more than 100% or actually undetermined; for the samples P-3 and P-3A it is 12.0%. Obviously these large errors in samples P-1 and P-2 are a result of the non-symmetry of these specimens.

Combining the errors, it is estimated that the k/ϵ values are accurate within 10% for the ZT type graphite (samples G-5 and G-9) and within 12% for the pyrolytic graphite (samples P-3 and P-3A).

Brown, et al.⁶ showed a steep increase in the room temperature thermal conductivity of graphites with an increase in their density. Considering the high density of the ZT type graphite, the values obtained for the thermal conductivities are in good agreement with the published data^{6,7,8,9,10} for similar types of graphite.

Comparison of the thermal conductivities of pyrolytic graphite is more difficult since their thermal conductivities depend upon many factors, especially the temperature of the deposition process⁷ and the treatment of the samples¹¹. Since all these factors are not available, an exact comparison is impossible although in general the measured values (for samples P-3 and P-3A) are in good agreement with earlier reported data^{11,12}.

The thermal conductivities of pyrolytic graphite are

reported here only at one temperature (1817°K) since the material is damaged when it is heated to higher temperatures; the samples split perpendicular to the z axis, and thus the measured temperature differences do not obey equation (4) any more. Such a structural change can be observed easily.

TABLE I

Radial Temperature Gradients

ZT Type Graphite, G-5 sample $2L = 0.622$ cm $2a = 2.537$ cm $T_0 = 1647^\circ\text{K}$

$$\Delta T_r = T(r, L) - T(r, 0)$$

at

$r = \frac{a}{4}$ r_{OF}	$r = \frac{a}{3}$ r_{OF}	$r = \frac{a}{2}$ r_{OF}	$r = \frac{2a}{3}$ r_{OF}	$r = \frac{3a}{4}$ r_{OF}	$r = a$ r_{OF}
2 +	7 *	21 +	39 *	48 +	66 +
2 +	8 *	19 +	36 *	45 +	65 +
4 +	5 *	20 +	32 *	46 +	61 +
8 ++	5 **	23 ++	38 **	49 ++	61 ++
3 ++	5 **	22 ++	36 **	44 ++	68 ++
8 ++	6 **	21 ++	40 **	46 ++	63 ++
5 ++					67 *
					63 *
					66 *
					67 **
					66 **
					67 **

$$\Delta T = 71.4^\circ\text{F} = 39.6^\circ\text{K}$$

+ ++ denotes one diameter, + on one side of center, ++ on other side of center

* ** denotes one diameter, * on one side of center, ** on other side of center.

The two diameters are perpendicular.

TABLE II

Radial Temperature Gradients

Pyrolytic Graphite, P-3 sample
 $2L = 0.378 \text{ cm}$ $2a = 2.314 \text{ cm}$ $T_0 = 1817^\circ\text{K}$

$$\Delta T_r = T(r, L) - T(r, 0)$$

at

$r = a/4$ r_{OF}	$r = a/3$ r_{OF}	$r = a/2$ r_{OF}	$r = 2a/3$ r_{OF}	$r = 3a/4$ r_{OF}	$r = a$ r_{OF}
1 +	10 *	12 +	38 *	37 +	82 +
3 +	12 *	12 +	27 *	33 +	73 +
4 +	12 *	13 +	41 *	36 +	74 +
0 +	10 *	16 +	40 *	42 +	70 +
4 ++	13 **	24 ++	35 **	55 ++	101 ++
12 ++	7 **	27 ++	32 **	57 ++	103 ++
5 ++	9 **	28 ++	30 **	66 ++	102 ++
7 ++	8 **	26 ++	32 **	57 ++	102 ++
					88 *
					84 *
					84 *
					82 *
					90 **
					84 **
					98 **
					96 **

$$\Delta T = 86.7^\circ\text{F} = 48.3^\circ\text{K}$$

TABLE III

Thermal Conductivity of ZT Type Graphite

$$2L_{G-3A} = 1.126 \text{ cm}, 2a_{G-3A} = 2.537 \text{ cm}$$

$$2L_{G-7} = 0.287 \text{ cm}, 2a_{G-7} = 2.539 \text{ cm}$$

$$2L_{G-5} = 0.622 \text{ cm}, 2a_{G-5} = 2.537 \text{ cm}$$

$$2L_{G-9} = 0.289 \text{ cm}, 2a_{G-9} = 2.538 \text{ cm}$$

Sample	T_o °K	ΔT °K	k_z/k_r	k_r/ϵ Cal/cm-sec-°K	k_z/ϵ Cal/cm-sec-°K	Gray Body Assumption $\frac{k_r}{\text{Cal/cm-sec-°K}}$ $\frac{k_z}{\text{Cal/cm-sec-°K}}$	
G-3A G-7	1671	31.8 73.0	0.255	0.405	0.103	0.306	0.0778
G-5 G-9	1260	12.0 20.0	0.153	0.488	0.0746	0.376	0.0575
G-5 G-9	1387	17.2 29.9	0.177	0.478	0.0848	0.366	0.0650
G-5 G-9	1647	39.6 72.0	0.213	0.395	0.0842	0.299	0.0638
G-5 G-9	2199	154.0 298.4	0.331	0.296	0.0980	0.218	0.0724

For (G₅-G₉)

$$k_z/k_r = (k_z/\epsilon)/(k_r/\epsilon) = -0.10116 + 2.00191 \times 10^{-4} \times T$$

Av. S.D. = 5.5%

$$k_r/\epsilon \text{ (Cal/cm-sec-°K)} = 0.61064 - 1.85699 \times 10^{-4} \times T$$

Av. S.D. = 5.0%

$$1260 < T < 2199^\circ\text{K}$$

TABLE IV

Thermal Conductivity of Pyrolytic Graphite

$$2L_{P-1} = 0.238 \text{ cm}, 2a_{P-1} = 2.540 \text{ cm}$$

$$2L_{P-2} = 0.163 \text{ cm}, 2a_{P-2} = 2.540 \text{ cm}$$

$$2L_{P-3} = 0.378 \text{ cm}, 2a_{P-3} = 2.314 \text{ cm}$$

$$2L_{P-3A} = 0.105 \text{ cm}, 2a_{P-3A} = 2.305 \text{ cm}$$

Sample	T_o °K	ΔT °K	k_z/k_r	Gray Body Assumption		k_r Cal/cm-sec-°K	k_z Cal/cm-sec-°K
				k_r/ϵ	k_z/ϵ		
P-1	1808	24.4	0.0636	2.579	0.0636	0.678	0.0167
P-2		30.5					
P-3	1817	48.3	0.0376	0.907	0.0341	0.238	0.0090
P-3A		105.6					

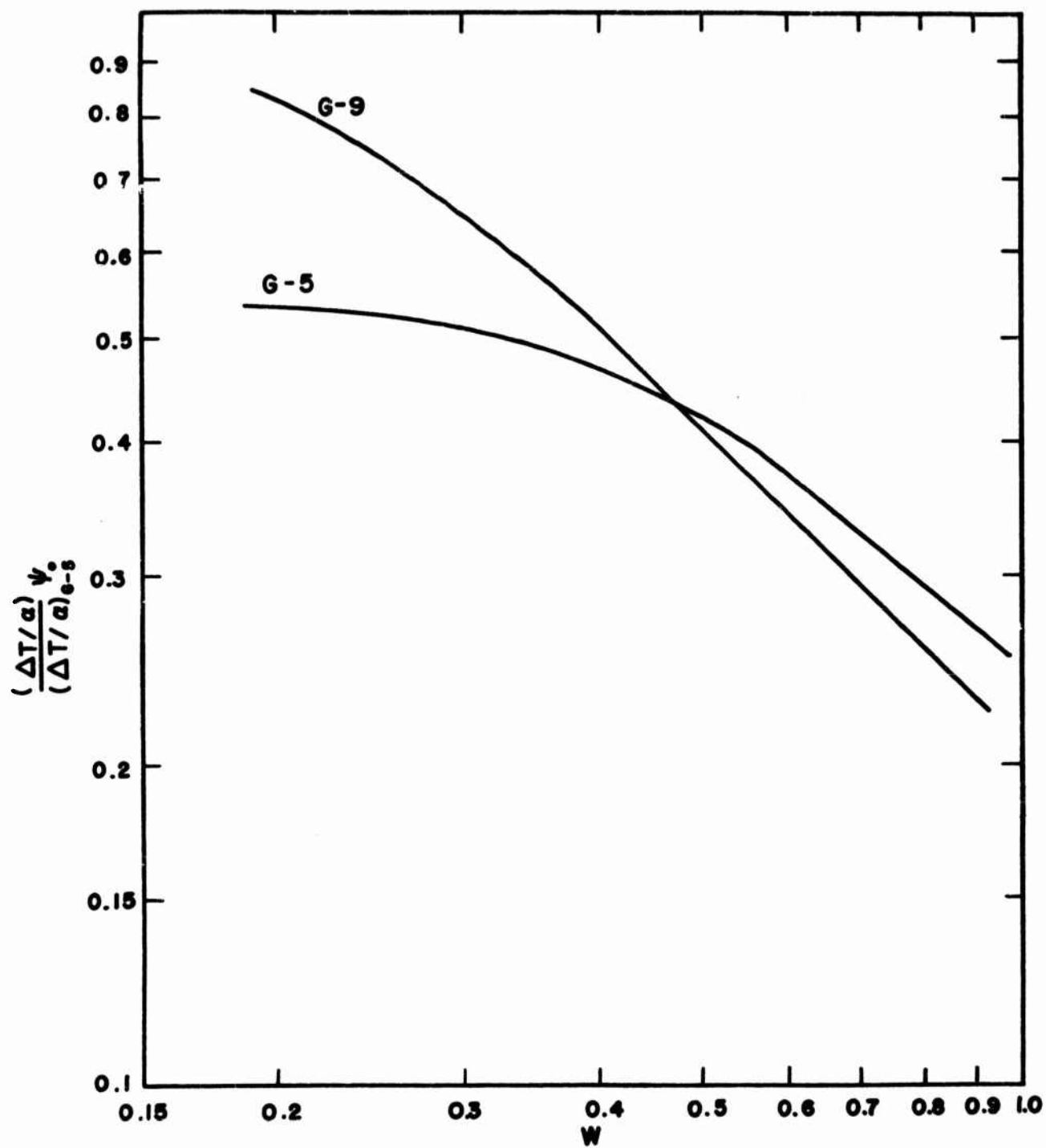


FIGURE 1: CALCULATION OF THE THERMAL CONDUCTIVITIES

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